



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

Docket No.: Q76184

Shunichi ISHIKAWA

Appln. No.: 10/606,236

Group Art Unit: 1772

Confirmation No.: 4220

Examiner: Sow Fun HON

Filed: June 26, 2003

For: GAS BARRIER FILM

DECLARATION UNDER 37 CFR 1.132

Honorable Commissioner of Patents and Trademarks,

Sir:

I, Hiroshi IWANAGA, a Japanese citizen, having a post office address of c/o Fuji Photo Film Co., Ltd., No.210, Nakanuma Minami-ashigara-shi, Kanagawa 250-0193 Japan, hereby declare and state that I received a Master's Degree from Tokyo Institute of Technology, Graduate School of Science and Engineering, Course of Polymer Engineering in March of 1990, and I was employed by Fuji Photo Film Co., Ltd. in April of 1990. I had been principally engaged in research and development of diffusion transfer photographic material from April of 1990 to August of 2000, nano particle materials from August of 2000 to November of 2002, organic-inorganic hybrid materials from November of 2002 to January of 2004, and organic and inorganic thin film materials from January of 2004 to date.

I declare further that I am the inventor of the subject matter of the claims in the above-identified application and I have read all of the

documents contained in the file wrapper of the above-entitled application.

I declare further that the test described below was conducted at my direction and under my supervision and the test results are true and correct to the best of my knowledge.

#### 1. Preparation of Materials

##### (1-1) Organophilized Layered Silicate C

5kg of montmorillonite (KUNIPIA, Kunimine Industries Co., Ltd.) as a layered silicate were added to 50 liters of mixture of water and methanol, and dispersed. Then, 5 kg of trimethyl hexadecyl phosphonium were added and the resultant mixture was stirred to obtain a layered silicate organophilized by trimethyl hexadecyl phosphonium ion (Organophilized Layered Silicate C).

##### (1-2) Organophilized Layered Silicate D

Organophilized Layered Silicate D was prepared in the same manner as Organophilized Layered Silicate C except that the same amount of distearyldimethylammonium chloride (DSDM) was used as an organophilizing agent instead of trimethyl hexadecyl phosphonium.

##### (1-3) Polyester Copolymer C

Condensation polymerization was conducted in the polymerization ratio of 45 mol % of 2,6-dinaphthalene carboxylic acid, 45 mol % of ethylene glycol, 5 mol % of o-aminobenzyl alcohol and 5 mol % of dimethyl 5-sodiumsulfoisophthalate to obtain Polyester Copolymer C.

##### (1-4) Comparative Polyester Copolymer C

Comparative Polyester Copolymer C was prepared in the same manner as Polyester Copolymer C except that terephthalic acid was used instead of 2,6-dinaphthalene carboxylic acid.

## 2. Preparation of Base Films

### (2-1) Film C

39.5 parts of Polyester Copolymer C, 10 parts of Organophilized Layered Silicate C, and 0.25 parts each of Irgafos 168 and Irganox 1010 (Ciba Specialty Chemicals) as antioxidants were added to 50 parts of molten polyethylene-2,6-naphthalate polymer at 300 °C. The mixture was kneaded and extruded by using a double screw extruder (Rheomix 60 OP/PTW25, Haake, Germany) to obtain Film C having a thickness of 200  $\mu\text{m}$ . Film C has a glass transition temperature of 125 °C and a linear thermal expansion coefficient of 10 ppm/°C.

### (2-2) Film D

Film D was prepared in the same manner as Film C except that Organophilized Layered Silicate D was used instead of Organophilized Layered Silicate C. Film D has a glass transition temperature of 114°C and a linear thermal expansion coefficient of 14 ppm/°C.

### (2-3) Comparative Film C

Comparative Film C was prepared in the same manner as Film C except that polyethylene terephthalate and Comparative Polyester Copolymer C were used instead of polyethylene-2,6-naphthalate polymer and Polyester Copolymer C, respectively, and the processed temperature was changed from 300 °C to 280 °C. Comparative Film C has a glass transition temperature of 78 °C and a linear thermal expansion coefficient of 14 ppm/°C.

### (2-4) Comparative Film C'

Comparative Film C' was prepared in the same manner as Comparative Film C except that the same amount of synthetic fluorine tetrasilicon mica (SOMASIF MTE, CO-OP Chemical Co., Ltd.) was used as an organophilized layered silicate instead of Organophilized Layered Silicate C. Comparative Film C' has a glass transition temperature of

81 °C and a linear thermal expansion coefficient of 12 ppm/°C.

### 3. Preparation of Gas Barrier Films

An inorganic coating layer and an organic-inorganic hybrid coating layer were formed on each of Film C and Film D in the same manner set forth in Example 1 of the present application to obtain gas barrier films (Sample C and Sample D).

The same process was tried to conducted on Comparative Film C and Comparative Film C' but wrinkles were generated during drying step at 120 °C. The drying condition was changed to 80 °C for 60 minutes and gas barrier films (Comparative Sample C and Comparative Sample C') were prepared from Comparative Film C and Comparative Film C' with the other conditions being unchanged.

### 4. Evaluation of Film Samples

The gas transmission rates of the gas barrier films produced above were measured by the MOCON method. The oxygen transmission rate was measured under conditions of 23°C and 0% of relative humidity. The water vapor transmission rate was measured under conditions of 23 °C and 90% of relative humidity. These conditions for the measurements are the same as Test Example 1 of the present application.

The results are shown in the following table. The table also shows the rest result of Sample B and Comparative B' described in Table 1 of the present application.

Sample	Base film		Test result	
	T <sub>g</sub> (°C)	CTE (ppm/°C)	Oxygen transmission rate (ml/m <sup>2</sup> ·day·atm)	Water vapor transmission rate (ml/m <sup>2</sup> ·day·atm)
B	168	35	0.02 or lower	0.02 or lower
C	125	10	0.02 or lower	0.02 or lower
D	114	14	0.02 or lower	0.02 or lower
Comparative B	163	60	0.04	0.04
Comparative C	78	14	0.05	0.04
Comparative C'	81	12	0.06	0.04

CTE: Linear thermal expansion coefficient

## 5. Discussion

The above table indicates that excellent gas barrier property can be attained only when (1) the gas barrier film has a base film containing an inorganic layered compound, (2) the glass transition temperature of the base film is 100 °C or higher, and (3) a linear thermal expansion coefficient of the base film is 40 ppm/°C or lower.

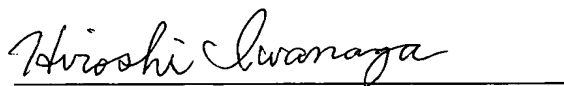
Pinnavaia et al., U.S. Patent No. 6,414,069 disclose inorganic layered compounds. However, Pinnavaia et al. merely mention that their inorganic layered compounds are useful as several kinds of agents including a barrier film agent (column 10, line 40-45) and do not describe a process for producing a barrier film. Pinnavaia et al. do not suggest how to attain the excellent barrier property of the barrier film. The excellent gas barrier property of the claimed invention cannot be attained by simply incorporating an inorganic layered compound into the base film since Comparative Samples C and C' comprising a inorganic layered compound in their base film exhibit inferior gas barrier property.

Matsuo et al., U.S. Patent No. 5,645,923 only disclose gas barrier films having a polyethylene terephthalate base film as working examples. Matsuo et al. are silent of inorganic layered compounds. If Pinnavaia et al. were combined with Matsuo et al., the excellent gas barrier property of the claimed invention cannot be attained by simply using a polyethylene terephthalate base film containing an inorganic layered compound. Please note that Comparative Samples C and C' having a polyethylene terephthalate base film containing an inorganic layered compound exhibit inferior gas barrier property.

Thus, I believe that the claimed invention is patentable over the cited references.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application of any patent issuing thereon.

Dated this 18<sup>th</sup> day of January 2006.

A handwritten signature in cursive script, reading "Hiroshi Iwanaga", is written over a horizontal line.

Hiroshi IWANAGA